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USASRDL Technical Report 2285

THE BEILBY LAYER

Walter J. Warden

July 1962

U. S. ARMY SIGNAL RESEARCH AND DEVELOPMENT LABORATORY
FORT MONMOUTH, NEW JERSEY

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Abstract

Consideration is given to experiments supporting the Beilby Layer Theory, i.e., an amorphous superficial structure can occur on the surface of a material as a result of a plastic flow developed by a polishing operation. This report is intended to illustrate the theory and stimulate interest in it because of its importance to the design, fabrication, and application of low-friction metallic items, glass lenses and prisms, semiconductors, and quartz frequency-control crystals.

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THE BEILBY LAYER

INTRODUCTION

Sir George Beilby, F. R. S., delivered a lecture before the Royal Society of London, England, on 21 November, 1907, in which he advanced a theory which has become known as the Beilby Layer Theory. Briefly, the Beilby Layer Theory states that when a solid, crystalline material is polished, a plastic flow can occur and create an amorphous superficial structure on the material. The depth of the layer is dependent upon polishing time, polishing materials used, and polishing techniques employed.* The opposite of the Beilby Layer Theory is stated simply in the belief that a polished surface is a mere reduction of surface roughness to such a degree that the finish of the surface is so extremely fine that for all intents and purposes the surface is completely smooth and lustrous. A middle-of-the-road belief is that an amorphous layer may be developed during polishing but it would be negligible and hence unimportant.

An understanding of the nature of a polished surface can be important for many reasons. In our highly industrialized age precision equipment is very important. Ball bearings which require highly polished surfaces are widely used in many types of precision equipments and are a vital part of virtually every mechanized industry. In time of war, the manufacture of ball bearings is a critical industry and considerable attention is given to locating and destroying the enemy's ball bearing manufacturing facilities. And the general subject of friction is a continuing, vital subject of study. Friction is the major cause of metallic wear which could very possibly be greatly reduced if the surface structures of polished materials were completely understood. At a meeting entitled "A Discussion on Friction" held in England on 19 April 1951, F. P. Bowden, F. R. S., of the Laboratory of Physical Chemistry, University of Cambridge, stated that the discussion fell into three main parts: "First, the friction of metals–about this we are beginning to know a little. Secondly, the friction of non-metals–about this we know far less, and I hope the discussion may focus attention of this fact. Thirdly, on boundary lubrication, where solids are covered with a few molecular layers of a lubricant."[1] And an industry which has progressed greatly since World War II is the manufacture of quartz (SiO_2) crystals. These crystals are used to stabilize electronic frequencies to ever-increasing accuracies. Every soldier of the past war who carried a walkie-talkie depended upon the successful operation of its crystal. E. A. Gerber and H. P. Wasshausen of the United States Army Signal Research and Development Laboratory reported, "...optical polishing has been used for finishing thin crystal plates and,... high-frequency crystals with polished and flat surfaces are far superior to those finished by common lapping techniques, especially at frequencies higher than 50 Mc."[2] Finally, it may be noted that manufacturers of optical equipment experience much waste while making mirrors, lenses, etc. for such equipment. A more thorough understanding of the surface structures of polished materials and the manner by which such surface structures are produced could lead to more efficient polishing equipment, materials, and techniques.

It is hoped that this report will encourage people to give more consideration to the nature and development of polished surfaces. The author believes that Beilby was at least very close to the truth, but also that there is plenty of room for an opposing theory. Such a theory could well begin a controversy which could ultimately lead to a full knowledge of the subject.

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*It must be noted that the Beilby Layer Theory does not completely discount any possibility of obtaining polished surfaces by other means. Rather, it emphasizes that a plastic flow can occur and does occur in most polishing operations.

There is not room to present all available evidence which supports the Beilby Layer Theory, but the following highlights should serve to explain it and possibly encourage more work to be done on its behalf.

EXPERIMENTAL DATA

Sir George Beilby, F. R. S., reasoned that the polishing of a solid, crystalline material could produce an amorphous layer upon the surface of the material. In his attempt to formulate such a polishing theory which has since come to bear his name—the Beilby Layer Theory—he performed, among many experiments, an experiment using speculum metal, an experiment using antimony, and an experiment using glass.

Speculum metal is an alloy of copper and tin. It is extremely hard and brittle and cannot be worked by ordinary tools. But it can be polished very easily, which Beilby did by using rouged leather (probably a polishing mixture of ferrous oxide and water used with a leather applicator).

Beilby first ploughed furrows into a piece of speculum metal with a watchmaker's file. He then examined photographs (1500 X) of the metal and there appeared to be a varnish-like effect over the surface of the furrows. He then worked the surface of the metal with emery paper of the finest grit obtainable. The file marks gave way to the many finer furrows made by the emery. Each such furrow was estimated to be 0.0003-inch wide. A very fine needle was then used to produce a scratch across the emery marks and it was estimated to be approximately 0.0005-inch wide. Reworking with the same emery paper resulted in small particles being lodged in the needle scratch. Another high-magnification photograph revealed that these particles were rounded and seemed more like drops than shavings. This indicated to Beilby that a plastic flow had occurred which resulted in the particles becoming rounded because of surface tension. Beilby concluded that the roundness of the particles indicated a flow had taken place on at least the outer portions of the particles.

The final steps were to polish the surface, remove the polished surface, and to repolish the surface. Beilby first polished the speculum metal with rouged leather. A photographic examination of the surface revealed, "The emery furrows have now completely flowed away, and a smooth layer has spread over the whole surface".[3] He then treated the surface with potassium cyanide which removed the polish and revealed the original crystalline structure. He then restored the surface by repolishing, and it was restored to the same appearance it had when first polished.

Beilby then experimented with antimony—a very brittle and fragile metal. Using the same methods he used with speculum metal, he produced furrows and pits on the surface of the antimony and then proceeded to polish the surface with rouged leather. Examinations of the surface revealed the same conditions as were present with the speculum metal. Beilby then stated, "In my own case... we must take it as established that the surface layer had for a brief period been in the condition of a viscous-liquid. Furthermore,... this flowed film is to be regarded as in a perfectly distinct state or condition from the original substance."*

Beilby also experimented with glass. He stated, "The existence of a definite skin on fire-glazed glass can be shown by lightly etching the surface with hydrofluoric acid vapour diluted with air. The surface is only slightly dimmed by this treatment, but the microscope

*Beilby's claim does not state that this flowed film can never be returned to crystalline structure. He indicates only that whether or not it does return to crystalline structure, there is a marked distinction between it and the original substance. He described the polished layer as being similar to an elastic stretched across the original surface.

shows that the vitreous reflecting surface has been completely removed, disclosing a finely granulated structure immediately below that surface." He scratched a piece of glass with a diamond and then worked the surface with emery. Although some flow was apparent, it was of negligible quantity to produce a polish. But when he worked the surface with wet rouge applied to hard wood rubber (sic), he produced, in a short time, a good polish. A photographic examination satisfied him that all surface irregularities had been covered over with a flowed surface layer.

After finishing the above experiments, Beilby concluded: "While the finest emery particles undoubtedly cause flow as they plough through the surface layer, yet the depth to which they penetrate disturbs the under surface and impairs the smoothness of the final surface layer. The rouge particles, it may be supposed, hardly penetrate below the surface, but coming into almost molecular contact with the sheet of molecules on the surface, drag it off like a skin. The fresh molecular layer left by the removal of the skin retains its mobility for an instant, and before solidification is smoothed over by the action of surface tension, thus producing the liquid-like surface which is the necessary condition of a perfect polish."*

Finch, Quarrell, and Roebuck of the Imperial College of Science and Technology, England, performed some experiments which were very pertinent to the Beilby Layer Theory. Their experiments consisted in observing the development of electron diffraction patterns during condensation of metal vapors on substrates. The results of their work seemed to them to "... confer an objective reality upon the Beilby Layer, which raises its existence from the realm of hypothesis to that of established fact."[4]

These three men discovered that they could not obtain ordered diffraction patterns of well polished surfaces. But when those surfaces were etched to remove the polish, characteristic patterns of the normal crystalline surfaces of the materials were obtainable. These men also found that metal films deposited on polished surfaces did not display diffraction patterns characteristic of their normal structure but such deposits did display their usual structure on unpolished substrates. Examination of diffraction patterns on polished surfaces, immediately following deposits of crystalline films, revealed that obtainable diffraction patterns of the deposits faded away as if the deposits were going into solution. The rates at which these patterns faded were determined largely by the methods employed to produce the polish.

Finch, Quarrell, and Roebuck agreed that suitably polished surfaces gave rise to random diffraction patterns; that etched materials exhibited crystalline structure patterns; and that patterns of crystalline deposits disappeared on polished substrates but remained on etched surfaces. They concluded that the experimental facts derived from their work offered direct proof of the existence of the Beilby Layer and its formation by polishing.

The work of notable scientists can often be enhanced by the endorsement of such work by capable, well-known, reputable colleagues. Such an endorsement was given to the work of Finch, Quarrell, and Roebuck (above) by Neil Kensington Adams, Sc. D., F. R. S. Adam's book, "The Physics and Chemistry of Surfaces" (1st ed. 1930), has long been considered an outstanding work regarding surface properties of materials.[5] His acceptance of the work of Finch, Quarrell, and Roebuck gives credence and support to their conclusions.

Adam also reported the work of Bowden regarding the relationship of melting points of polishers and substances being polished. Bowden's work showed, both theoretically and experimentally, that the surface temperature of a solid during friction can rise quickly to its melting

*Beilby defined a perfect polish as, "... one which perfectly reflects a beam of light according to the well-known law of reflection; if any stray light leaves the surface at angles other than that of the incident beam, the polish is imperfect. A beam of light falling on the clean, undisturbed surface of a liquid is reflected in this way..."

point but *never rises higher than this,* and that polishing occurs only if the melting point of the polisher is higher than the melting point of the substance being polished. Hence, should two materials be made to act one upon the other, polishing, with rare exception, will occur on the surface of the material which possesses the lowest melting point. Bowden showed that oxamide (m.p. 417°C) will not polish speculum metal (m.p. 745°C) but speculum metal can be polished by lead oxide (m.p. 888°C). Calcite (m.p. 1339°C) can be polished by stannous oxide (1625°C) or by zinc oxide (m.p. 1800°C), but not by cuprous oxide (1235°C).

Bowden's work indicated that hardness is of little importance in polishing. But a few cases were found where very ductile metals such as gold and platinum could be polished with materials of melting points far below that of the metals.

Adam's studies of the work of Finch, Quarrell, and Roebuck and Bowden and others led him to write, "... the amorphous Beilby Layer Theory has properties markedly different from the rest of the solid." Elsewhere, he wrote, "The various forms of cold-working of metals all seem to produce a more or less perfect Beilby layer on the surface, with consequent increase in hardness and *resistance to mechanical wear.*" (Italics added)

There are hundreds of ways to produce polishes. Lapidaries follow one or both of two generalized systems in determining how best to polish the materials with which they are concerned. One system is to devise by hit-and-miss methods their own combinations which react best on a particular material. The other system is to follow the advice of others who have discovered (usually by hit-and-miss methods) some way of suitably working various materials. As sources of information for the latter system, a few textbooks are available, which offer information concerning successful work of others.

One such book was written by F. Twyman, F. R. S., entitled "Prism and Lens Making."[6] In it, he describes a particularly interesting method which was used for some time in France to dry-polish glass. Letter paper sold in Paris under the name "Berzelius paper" was cemented onto a lap and after the cement was sufficiently dry the papered lap was then sprinkled with powdered tripoli. Glass was then polished in the usual way but fine scratches were very likely to develop. However, the scratches were sufficiently reduced by removing superfluous powder from the lap, breathing upon the lap, and then drawing the glass across the lap while using heavy pressure.

The above dry-polishing method indicates several factors. First, tripoli powder is harder than glass because of its ability to scratch the glass. Secondly, in view of the work of Bowden (above) the melting point of tripoli is very likely higher than that of the polished glass. Thirdly, the addition of the soft background of paper could allow for a reduction of the abrading ability of the tripoli particles.

In view of these factors an assumption is made that this method of dry-polishing glass lends support to the theory of the Beilby Layer development because of the great amount of heat which could be developed upon the surface of the glass while at the same time keeping the abrading ability of the polishing particles to a minimum. Either one of two things occurred which suitably reduced the original scratches to a point where an acceptable polish was obtained. Either a sufficient amount of glass was removed to reduce the surface to the level of the bottom of the scratches or the scratches were filled. There is no evidence to support the former possibility, especially in view of the fact that the amount of polishing material was reduced to a minimum. But the latter possibility appears plausible and commensurate with the Beilby Layer Theory.

Twyman also reported that lapidaries of Hilger & Watts, England, customarily dry-polish copper on greyed quartz. Here again the polisher, although harder than the material being polished, has a higher melting point than the material being polished. And in this case there is no

loose compound or mixture acting as a polishing or abrading agent. Hence, consideration must again be given to the development of a Beilby layer.

Twyman's text also makes reference to a Hilger & Watts method of polishing synthetic ruby (a form of corundum) by the use of a flame. In this case it is quite obvious that heat is the only factor involved which lends further support to the Beilby Layer Theory.

Two Japanese researchers, Nonaka and Kohra, reported in 1954 some interesting results of work performed with polished surfaces of copper, nickel, and gold. Although they were reluctant to accept, in toto, the Beilby Layer Theory, they did join the slowly growing number of men convinced that a polished surface of a material is different from the original material. In reference to the polished layer, they wrote, " We mean by it the polished layer of metal which has a mirror-like appearance and gives a diffraction pattern of diffuse haloes."[7] But throughout their report they continued to refer to this layer as the Beilby layer.

Their experiments were directed toward a study of the recrystallization of polished surfaces and the recentness of their work indicates that much remains to be learned of the nature of polished surfaces of various materials. Their studies of Beilby layers showed that a definite change occurred on a metal surface after careful polishing. The new surface, however, was itself subject to change when exposed to various atmospheres or vacuum. Recrystallization of the layers as well as oxidation of the layers (except for gold which does not oxidize) occurred in varying degrees.

American scientists, on the whole, are apparently hesitant to give credence to the Beilby Layer Theory. It is not uncommon to hear an American scientist say, "What's that?" when queried about the Beilby layer.

But one American, Kamalesh Ray, reported[8] that an examination of spent rouge, which was used to polish glass, revealed the presence of some silica (glass particles), but not enough to account for the total polish. He stated, "Other experiments indicate that (a) "filling-up" process through softening of the glass surface is definitely involved, and calculations show that a sufficient amount of heat through polishing friction is available for the purpose." He felt that if rouge particles are harder than glass then scratches should occur on the surface of glass polished with rouge. However, examinations of rouge-polished glass at 8000 to 20,000 magnifications revealed no such scratches.*

Ray also reported an interesting observation (unpublished) made by Dr. J. A. Anderson, Astrophysics Department, California Institute of Technology, concerning the Mt. Wilson Observatory. Old lenses of the observatory contained very-minute surface cracks similar to those which had been reported on old telescope lenses in different parts of the world. It was felt that periodic heating and cooling over a period of years could have caused them. Dr. Anderson reproduced such surface cracks by laboratory methods on ordinary lenses and found them to be of the same order as the natural cracks.

Ray reports, "These observations suggest that the skin of a rouge-polished lens has physical properties different from the bulk of the lens body. This may be possible if the *polishing takes place through thermal (softening) action at the superficial* layer." **(Italics added)**

The author became quite interested in the Beilby Layer Theory and performed a few experiments. The first one was an attempt to polish a quartz crystal (approximately 0.006-inch

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*It should be noted that Ray speaks elsewhere of laps made with pitch, resin compound, felt, etc., so it is not known how the glass was polished.

thick and 0.450-inch diameter) with a sheet of platinum. The surface finish of the crystal had been prepared with 5-micron abrasive. The platinum was mounted in a lathe and made to revolve at several hundred revolutions per minute. The crystal was then pressed by a finger tip against the platinum and after a short time, a dull polish appeared at the center of the crystal. This was the first time, to the knowledge of the author, that a quartz crystal had ever been polished dry without the use of a loose polishing compound or polishing mixture.

A second experiment concerned flash-plating of an optically polished quartz crystal and a 5-micron-finish unpolished crystal. Thirty seconds of aluminum plating on each one resulted in plainly visible layers of aluminum. Both were rubbed with optical paper and while some plating was removed from the unpolished crystal none came off of the polished crystal. Attempts were then made to plate two more such crystals by subjecting them to no more than a few seconds of plating. Four attempts failed to produce visible layers of plating. On the fifth attempt, plating appeared on the polished crystal but not on the unpolished crystal. One-half to one hour later the plating on the polished crystal appeared (by eye) to have spread. Two hours after plating, the layer of aluminum seemed to be "gone." Approximately 2-3/4 hours after plating, a witness confirmed the fact that the plating had disappeared. This seemed to be in conformity with the work of Finch, Quarrell, and Roebuck (above) who reported crystalline deposits on polished surfaces fading away as if they were going into solution. The author fully realizes that minute films of aluminum oxide are invisible which could account for the disappearance. But aluminum is one of the fastest oxidizing of all metals and the time factor of nearly three hours tends to throw support to the solubility theory and hence to the Beilby Layer Theory.

A third experiment was a continuation of an experiment designed to study the results of etching quartz crystals. It was originally conducted some years ago by Warren Roberts, a geologist of USARSDL who has since transferred to another government installation. From a group of approximately 30 photographs estimated to be at least 5000X, taken of various crystals, only three show a direct sequence taken at exactly the same spot. Figures 1, 2, and 3 show a section of an optically polished quartz crystal with no etching, 1-minute etching, and 5-minute etching, respectively.

It is difficult to imagine that a beautiful, optically polished quartz crystal can possess such ugly deformities as are shown by the photographs. A simple examination of the photographs shows that etching removes what seems to be a very definite viscous layer spread across the deformities. The snake-like scratch which crosses the upper right-hand corner illustrates this very nicely and conforms precisely with the work of Beilby on speculum metal and antimony. Figure 3 shows that 5-minute etching reveals a very dark line within the snake-like scratch. This darkness is attributed to shadows formed within the scratch from the light used to illuminate the crystal while it was being photographed. Figure 1, no etching, does not show darkness in this scratch, thus indicating that the scratch was filled or covered over during the polishing operation.

In the opinion of the author, his preceding experiments support the Beilby Layer Theory. But it is fully realized that they represent hardly a beginning to what can and should be done in making an exhaustive study of the Beilby Layer.

Some interesting observations have been made about polishing by a leading lapidary, H. P. Wasshausen. He is formerly of Carl Zeiss, Jena, Germany, manufacturers of all kinds of optical equipment and quartz crystals. His experience has covered a period of more than 30 years. The following excerpts have been taken from many conversations between Mr. Wasshausen and the author:

1. A minimum amount of liquid in a polishing mixture will enable one to obtain the fastest polish. But too fast a polish is an indication of excessive heat which may cause distortion of quartz crystals.

2. For quick polishing, in instances where surface flatness is of no concern, a felt lap is used with high pressure and high speed.

3. Weight is a determining factor in polishing.

4. A polishing operation is apt to remove more in total thickness when the polishing plate is heavy.

5. Aluminum oxide may or may not scratch quartz crystals during polishing. (Scratching could depend upon purity of the compound or the selection of lap material—author's note.)

6. Ceric oxide polishes very fast but tends to make the corners of quartz blanks round. (Ceric oxide is pyrophoric (spark-emitting) which could account for excessive heat and thus undue polishing at the extremities of the blanks—author's note.)

7. New methods are being developed to use diamonds for polishing. (The use of diamonds in lapping requires laps which offer a soft background—author's note.)

One can find support for the Beilby Layer Theory in the statements of Mr. Wasshausen, although they are by no means intended to be offered as final proofs. An interesting aspect of the statements lie in the fact that technicians can be as vital as solid-state physicists for the gathering of evidence. Should the Beilby Layer Theory ever be proved or disproved, much work will have to be done by both such types of men.

APPLICATIONS

The functional qualities of low-friction metallic items, glass lenses and prisms, semiconductors, and quartz, frequency-control crystals continue to become increasingly more complex and meaningful. For example, quartz, frequency-control crystals are required to satisfy smaller tolerances and greater reliability than ever before. Increased attention is, therefore, being directed to all phases of their fabrication, including surface finish. A complete knowledge of surface finishes of quartz crystals would be helpful to their design, fabrication and applications.

CONCLUSIONS

It may be concluded, in view of the foregoing, that melting points of compounds and other materials can be significant in polishing operations. Table 1 shows melting points and hardness values for commonly used compounds; values were unobtainable for some materials. Such values may be important in future research on the Beilby layer. Hardness may or may not be an important quality, but brittleness, mentioned very little, could be important to polishing operations. Brittle materials, which are easily broken, could produce additional heat in the breaking-down process, and this heat could aid polishing. As yet tables of brittleness are not to be found.

THE AUTHOR SURMISES THAT AN IDEAL POLISHING AGENT TO BE USED FOR POLISHING A MATERIAL IS ONE THAT HAS A HIGHER MELTING POINT, A LOWER HARDNESS VALUE, AND YET IS MORE BRITTLE THAN THE MATERIAL TO BE POLISHED.

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TABLE I[1]

Material	Melting Point °C	Relative Hardness[2]	Mohs' Hardness
Diamond (C)	3500	1	10
Carborundum (SiC)	2600	2	9–10
Ceric Oxide (CeO_2)	2600	?	?
Magnesia (MgO)	2500–2800	9	4.8
Emery (Corundum) (Al_2O_3)	2050	3	9
Chromium Oxide (Cr_2O_3)	1990	?	?
Platinum (P_t)	1773.5	10	4.3
Cerous Oxide (Ce_2O_3)	1692	?	?
White Rouge[3] ($Ca3(PO_4)_2$)	1670	7	5–6
Red Rouge (Fe_2O_3)	1565	8	4–7
Quartz (SiO_2)	1470	4	7
Nickel (Ni)	1455	6	6
Putty Powder (SnO_2) (White amorphous powder)	1127	5	6–7
Copper (Cu)	1083	12	2.5–3
Speculum Metal (Alloy of copper and tin)	745[4]	?	?
Antimony (Sb)	630.5	11	3–3.3
Tripoli (Rotten Stone)[5]	?	?	?
Talc ($3M_gO \cdot 4SiO_2 \cdot H_2O$)	?	13	1

[1]Melting point and hardness values vary slightly in different reference books for some materials. These values, with the exception of the melting point for speculum metal, were obtained from the following two sources:

a. Hodgman, Charles D., Robert C. Weast, and Samuel M. Selby (eds.). Handbook of Chemistry and Physics, 41st ed., Chemical Rubber Publishing Co. (Cleveland, Ohio), 1959.

b. West, Clarence J., and Callie Hall (compilers). International Critical Tables of Numerical Data, Physics, Chemistry and Technology. McGraw-Hill Book Company, Inc. (New York and London), 1933.

[2]The relative hardness values are only for those materials in this table. The lowest value represents the hardest material, etc.

[3]White rouge may be one of many things. Suppliers of white rouge usually remain silent as regards its composition. Some people believe that white rouge is powdered quartz (SiO_2) while others believe it may be powdered aluminum oxide.

[4]This value was obtained from the following source: Adam, Neil Kensington. The Physics and Chemistry of Surfaces, 3rd ed., Oxford University Press (London), 1941, p. 173, 1. 21.

[5]Tripoli is a porous siliceous rock which, when pure, contains about 98% silica and generally some alumina and oxide of iron. It is found in many forms and in various parts of the world. Specific hardness and melting point values are unobtainable.

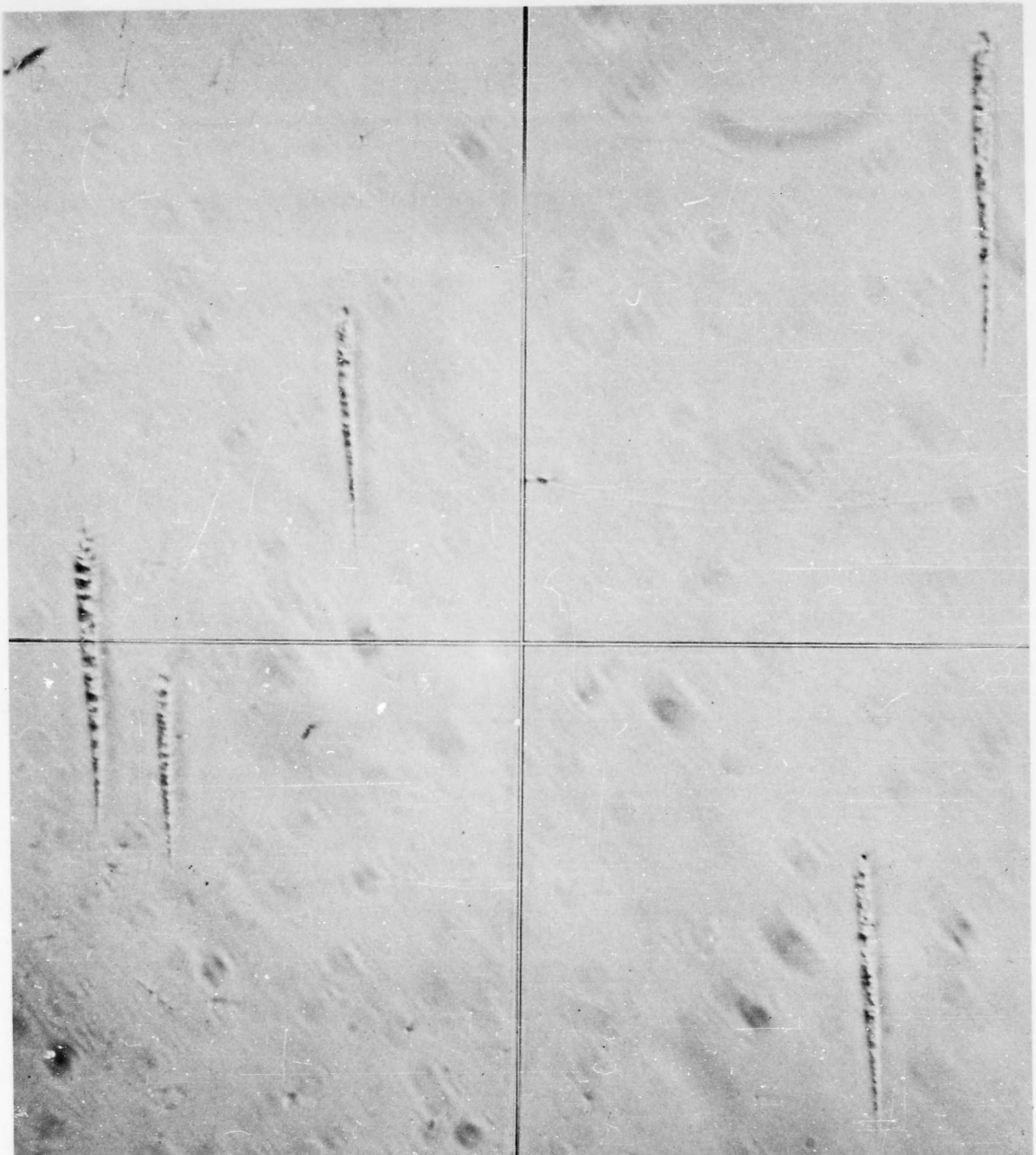

FIGURE I
OPTICAL POLISH
NO ETCH

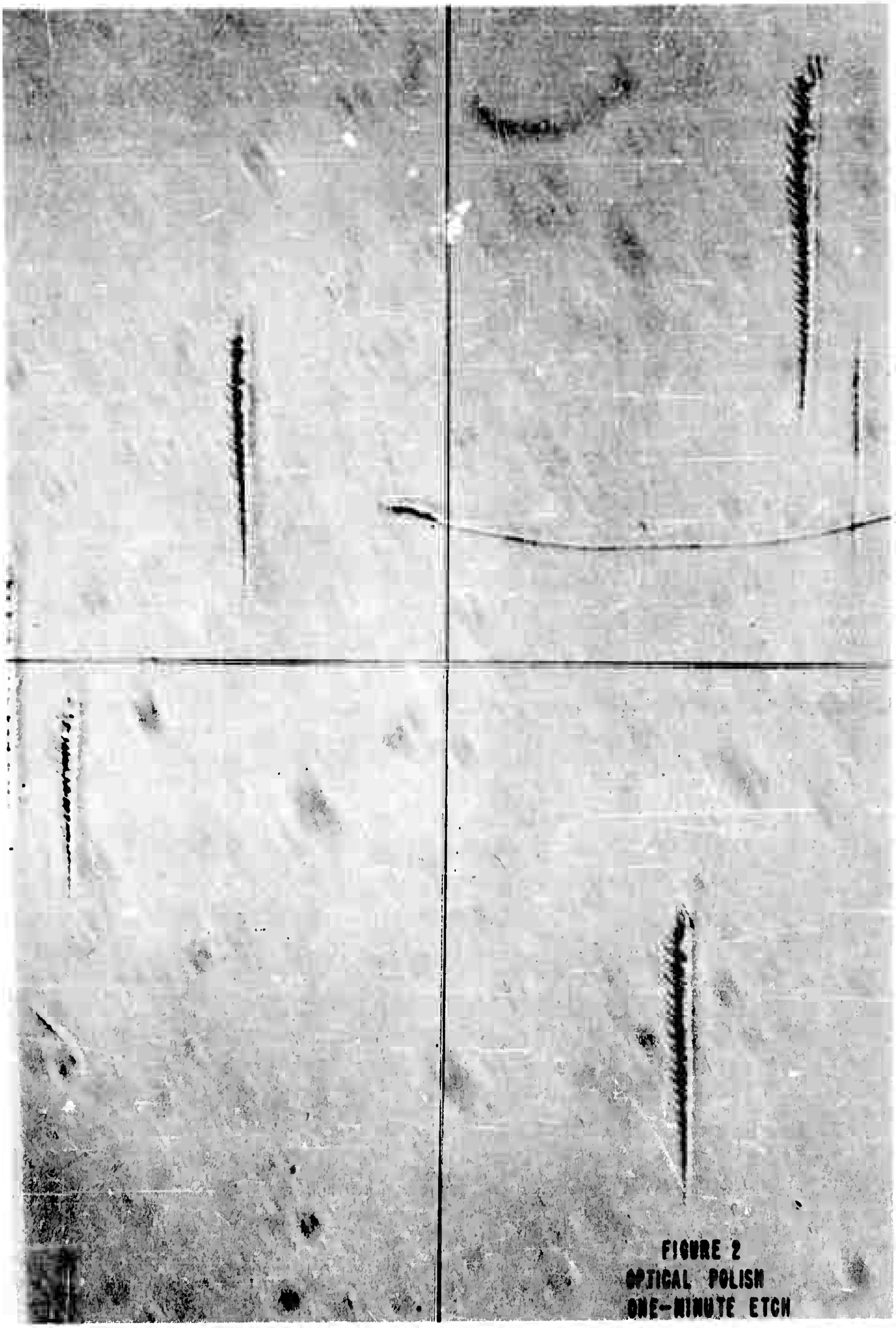

FIGURE 2
OPTICAL POLISH
ONE-MINUTE ETCH

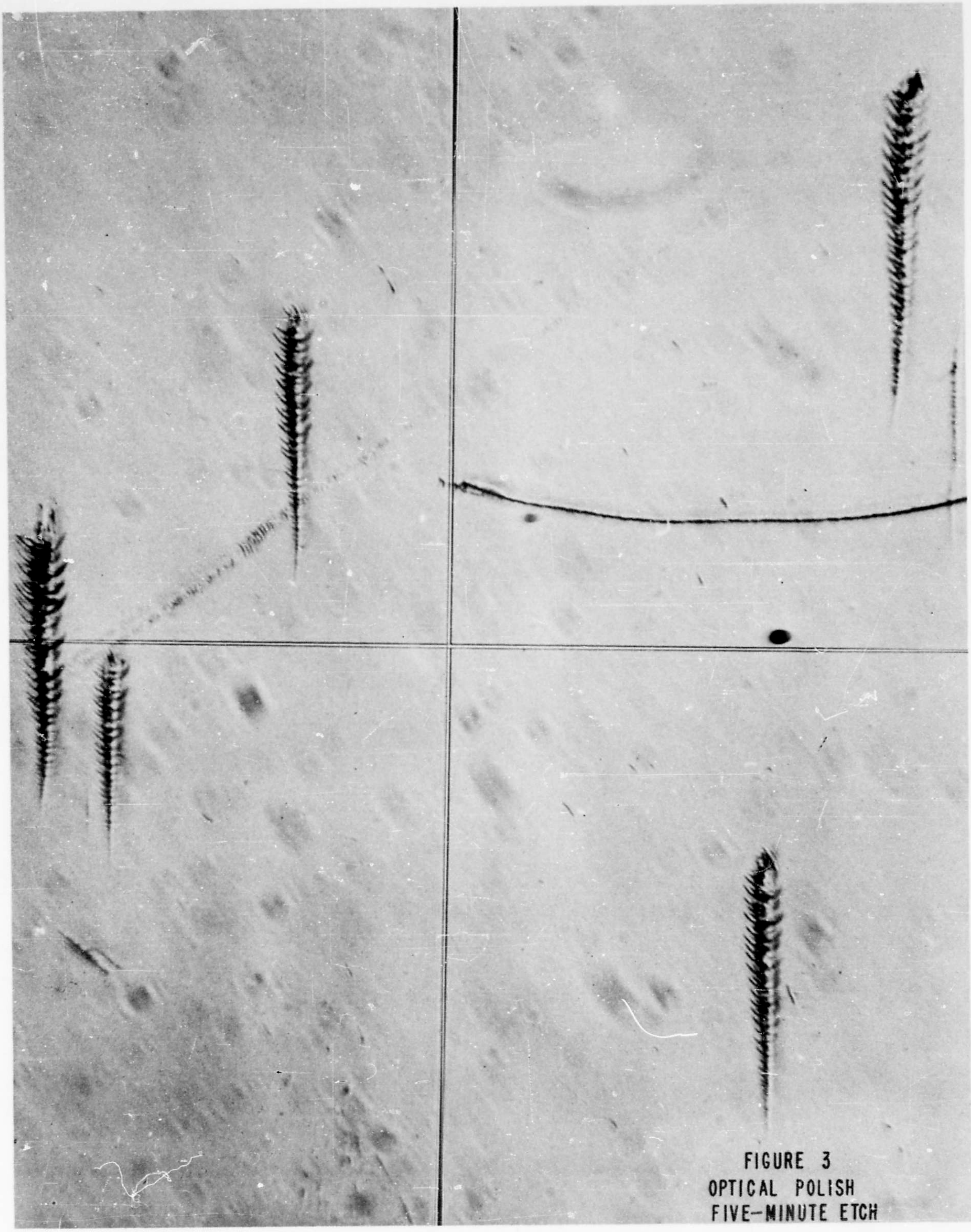

FIGURE 3
OPTICAL POLISH
FIVE-MINUTE ETCH

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Army Signal Research and Development Laboratory
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THE BEILBY LAYER by Walter J. Warden
June 1962, 12 p. incl. illus. table, 8 refs.
(USASRDL Technical Report 2285)
(DA Task 3A99-15-004-01) Unclassified Report

Consideration is given to experiments supporting the Beilby Layer Theory, i.e., an amorphous superficial structure can occur on the surface of a material as a result of a plastic flow developed by a polishing operation. This report is intended to illustrate the theory and stimulate interest in it because of its importance to the design, fabrication, and application of low-friction metallic items, glass lenses and prisms, semi-conductors, and quartz frequency-control crystals.

UNCLASSIFIED

1. Polishing
2. Plastic flow, Polishing
3. Amorphous structure, Polished surfaces

I. Warden, Walter J.
II. Army Signal Research and Development Laboratory, Fort Monmouth, N. J.
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